Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

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Public Comments

No public comments were received for this proposal.

Technical Synthesis Panel Review

Proposal Title

#0341: Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

Final Panel Rating
adequate

Technical Synthesis Panel (Primary) Review

TSP Primary Reviewer's Evaluation Summary And Rating:

The researchers propose a study to better understand transport of Hg in a shallow regions of the Bay−Delta system. Since Hg is transported mainly via particles, the researchers look in great detail at the processes they presume dictate Hg particle partitioning, intraparticle diffusive mass transfer and sorption−diffusion processes. They justify the study by acknowledging that most current models use simple portioning to describe particle−water interactions, without looking at kinetics of the processes and the intraparticle reactions. They are well qualified to conduct the mass transfer and hydrodynamic modeling for this type of study. Unfortunately, this proposal contains very little geochemistry, which is essential to understanding behavior of Hg in the system. Aqueous speciation of Hg is dominated by hydrophilic processes, yet many of the applications of mass transfer models have been developed for hydrophobic compounds such as PCB's and PAH's. Furthermore the study treats particles as if they were inert surfaces and an extensive body of literature has been published on reactivity of phytoplantonic surfaces. Similarly, processes occurring at the sediment−water interface would be highly dependent on diagenetic processes of degradation of recently−deposited planktonic biomass. This project would definitely benefit from the close alignment with

a Hg biogeochemist or perhaps developing the hydrodyanamic and kinetic portions of the model into existing models (EPRI's MCM model, for example). Reviewers also suggest a laboratory based study as a "proof of concept" approach prior to submitting a revised proposal.

Additional Comments:

The researchers propose a study to better understand transport of Hg in a shallow regions of the Bay−Delta system. Since Hg is transported mainly via particles, the researchers look in great detail at the processes they presume dictate Hg particle partitioning, intraparticle diffusive mass transfer and sorption−diffusion processes. They justify the study by acknowledging that most current models use simple portioning to describe particle−water interactions, without looking at kinetics of the processes and the intraparticle reactions. They are well qualified to conduct the mass transfer and hydrodynamic modeling for this type of study. Unfortunately, this proposal contains very little geochemistry, which is essential to understanding behavior of Hg in the system. Aqueous speciation of Hg is dominated by hydrophilic processes, yet many of the applications of mass transfer models have been developed for hydrophobic compounds such as PCB's and PAH's. Furthermore the study treats particles as if they were inert surfaces and an extensive body of literature has been published on reactivity of phytoplantonic surfaces. Similarly, processes occurring at the sediment−water interface would be highly dependent on diagenetic processes of degradation of recently−deposited planktonic biomass. This project would definitely benefit from the close alignment with a Hg biogeochemist or perhaps developing the hydrodyanamic and kinetic portions of the model into existing models (EPRI's MCM model, for example). Reviewers also suggest a laboratory based study as a "proof of concept" approach prior to submitting a revised proposal.

Technical Synthesis Panel (Discussion) Review

TSP Observations, Findings And Recommendations:

Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

The researches propose development of a complex mathematical model to describe the particle−associated fate and transport of mercury. The issue of intra−particle processes adds an interesting and potentially important new component to Hg transport and fate models. The proposal addresses an important issue of concern to CALFED, but the reviewers had difficulty in discerning how the model could be used effectively or whether it would be a useful tool for broader CBDA application, particularly in risk assessment. The model treats all particles equally and doesn’t address important aspects of Hg geochemistry and surface characteristics of planktonic particles and surface associated DOC. It is unclear how it could be effectively integrated with existing hydrodynamic models of Hg transfer without this level of detail. The proposal does not demonstrate how the model will be validated.

The panel recommended that the researchers develop a future proposal with Hg geochemists who are familiar with complexities of Hg geochemistry.

Final Ranking: Adequate

Technical Review #1

proposal title: Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

Review Form

Goals

Are the goals, objectives and hypotheses clearly stated and internally consistent? Is the idea timely and important?

Comments	**The goals for this project are well stated, extremely timely and important, and extraordinarily ambitious. The various tasks outlined are internally consistent and individually important.**
Rating	**excellent**

Justification

Is the study justified relative to existing knowledge? Is a conceptual model clearly stated in the proposal and does it explain the underlying basis for the proposed work? Is the selection of research, pilot or demonstration project, or a full−scale implementation project justified?

Comments	**Understanding the transportation and fate of mercury in aquatic environments is vitally important to a variety of problems worldwide and constitutes more than enough justification for the proposal. The heavy mercury loading in the San Francisco Bay as a result of 19th century hydraulic mining will be a problem for decades to come and any tools that can be made available to engineers and planners will be helpful. This proposal represents an important step in that direction.**
Rating	**excellent**

Approach

Is the approach well designed and appropriate for meeting the objectives of the project? Is the approach feasible? Are results likely to add to the base of knowledge? Is the project likely to generate novel information, methodology, or approaches? Will the information ultimately be useful to decision makers?

Comments	**The general approach to the project has a logical sequence and is fundamentally sound. The proposal does have particular strengths and weaknesses that merit detailed commentary. The PIs are clearly involved in cutting edge research on a variety of experimental and modeling techniques. All of these will be necessary to carry out the proposed project. The proposal has a number of laudable commendations. (1) Use of the Mellor−Yamada model for handling turbulent flow in hydrodynamic models has become the de facto standard for successful fluid modeling in complicated natural settings such as estuaries. (2) A large amount of previous experiments and modeling of mercury transport (e.g., Bale's work) exists and will be incorporated by the project scientists. Other approaches in the proposal appear weaker or are at least poorly explained. (1) The particle diffusion model described in Task 1 makes sense but lacks a strong track record of successful prediction. The successful results appear to consist of one chemical component and one substrate (top of p. 8, figure 5) and a manuscript in preparation. (2) It seems to me that the chemical composition of sediment particles would be highly variable but extraordinarily important in understanding diffusion rates. (Surely quartz is going to behave differently than particulate organic carbon). (3) The mechanics of the rate limiting steps (equations 2 and 3) are difficult for scientists not familiar with these concepts to comprehend. (4) Successful prediction of erosion of cohesive sediment is one of the most daunting tasks in the realm of hydraulic engineering. The models given by in equations (7) – (11) from the papers by Garcia and Parker (complicated though they may be) apply to much**

	simpler non−cohesive sediment transport. In the San Francisco Bay, the PIs will almost certainly be dealing with very complex, very fine−grained cohesive sediments.
Rating	good

Feasibility

Is the approach fully documented and technically feasible? What is the likelihood of success? Is the scale of the project consistent with the objectives and within the grasp of authors?

Comments	The feasibility of the project is possibly its weakest link. While the justification and goals of the project are excellent, the budget and timeframe necessary to carry out the very ambitious goals of the project seem too modest.
Rating	good

Monitoring

If applicable, is monitoring appropriately designed (pre−post comparisons; treatment−control comparisons)? Are there plans to interpret monitoring data or otherwise develop information?

Comments	The PIs appear to have made a good faith effort to validate the model using data from a relatively small subdomain of the San Francisco Bay.
Rating	very good

Products

Are products of value likely from the project? Are contributions to larger data management systems relevant and considered? Are interpretive (or interpretable) outcomes likely from the project?

Comments	The products produced by the project (journal articles and two scientific workshops) are modest but in line with the project goals and

	budget.
Rating	**very good**

Additional Comments

Comments

Capabilities

What is the track record of authors in terms of past performance? Is the project team qualified to efficiently and effectively implement the proposed project? Do they have available the infrastructure and other aspects of support necessary to accomplish the project?

Comments	**The PIs all appear to be capable scientists who can carry out the proposed work. The lead PI seems to be a bright and ambitious young research engineer.**
Rating	**excellent**

Budget

Is the budget reasonable and adequate for the work proposed?

Comments	**The budget for the project ($132,000) is very modest given the scope of the proposed work. My sense is the PIs are promising far more than they can deliver for the limited funding being requested, the short time frame of the project, and the weaknesses in the approaches given above.**
Rating	**very good**

Overall

Provide a brief explanation of your summary rating.

Comments	**This is a worthwhile and important proposal that deserves funding, if not in this cycle then certainly in the future. The PIs are overreaching in terms of**

	what can be delivered for the budget and time frame they propose. This constitutes the area where the proposal most needs to be strengthened. A more realistic proposal might involve the experimental and analytic work described in Tasks 1 and 2 (with correspondingly greater discussion of the details) plus some variation on Task 4 to field test the results in the sediments of Grizzly and Suisun Bays. The combined hydrodynamic−sediment transport model (all to be adapted to a new parallel processing scheme) is a laudable goal but it has not addressed the very difficult technical issue of modeling cohesive sediment transport.
Rating	**very good**

Technical Review #2

proposal title: Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

Review Form

Goals

Are the goals, objectives and hypotheses clearly stated and internally consistent? Is the idea timely and important?

Comments	**The goals are clearly stated and internally consistent. While the hypotheses to be tested are of potential importance, the proposal does not make a compelling argument that the proposed research is a critical step in the evolution of our understanding of mercury dynamics in coastal marine systems.**
Rating	**very good**

Justification

Is the study justified relative to existing knowledge? Is a conceptual model clearly stated in the proposal and does it explain the underlying basis for the proposed work? Is the selection of research, pilot or demonstration project, or a full−scale implementation project justified?

Comments	**The proposed modeling is reasonably well described in the proposal but there is little evidence of the potential significance of the work. For example the temporal framework of reversible intra−particle sorption kinetics are not presented. Initial estimates of the importance of the intra−particle dynamics with respect to overall mass transfer relative to surface interactions (sorption processes limited to the surface of the particles themselves) would have been quite useful.**

Presumably the intra particle dynamics would be much more important for larger radius particles than the smaller size particles with their larger surface to volume ratios, but the major mass of sorbed mercury are in the fine particle fractions.

The importance of intra−particle transport has been recognized for some time in describing the sorption process since equilibrium is not completely reached in short−term experiments. Nyfeller et al. (1984) recognized this and presented a simple two step kinetic model to describe the role of intra particle diffusion as a rate limiting step in reaching complete equilibrium in the sorption of metals by natural particles. Adsorption of the majority of the sorbed metals however occurred rapidly, with only minor fractions of the total sorbed species being slowly adsorbed thereafter. The slow adsorption of the minor fraction was considered to be limited by radial diffusive processes (and complicated by internal reactions and their kinetics during transport and interactions within the compositionally varying media within the particles themselves as shown by subsequent work). The process of radial diffusive mass transport of organics within natural particles has also been discussed in the popular textbook by Schwarzenbach et al. 1993.

In addition the desorption kinetics of mercury may be so slow that effective desorption rate time scales may be much longer than the time scale of short−term processes such as resuspension events to be addressed in the proposed model and thus be treatable as “inert” rather than as a kinetically labile species. Indeed release by early diagenetic processes may be of greater importance but

	unfortunately will not be addressed in the proposed model.
Rating	**fair**

Approach

Is the approach well designed and appropriate for meeting the objectives of the project? Is the approach feasible? Are results likely to add to the base of knowledge? Is the project likely to generate novel information, methodology, or approaches? Will the information ultimately be useful to decision makers?

Comments	**The proposed research argues that there is a need to include intra−particle transport in modeling the sorption of mercury by natural particles in the water column and sediment and proposes what appears to be a reasonable approach to incorporate this process in 3−D models already developed for Grizzly and Suisun Bays. The hypothesis that "a primary process controlling the concentration of mercury species including methyl mercury in the dissolved phase is the rate of mass transfer of mercury to and from the particulate phase; further we believe that this mass transfer is controlled by both sorption−desorption and diffusion within the pore spaces of particles" in the absence of even rough approximations seems quite tentative and not supported by existing data. Initial quantitative evaluation of the potential significance (e.g. need to include) the intra−particle diffusive process in describing mercury sorption on natural particles would have been most useful.** **Secondly there is no indication that they intend to validate the model using appropriate laboratory sorption−desorption kinetic studies before applying the model as**

	a subroutine in the 3−D model. They do, however, refer to studies fitting their model to the adsorption of 1,2−Dichlorobenzene to shale in which, incidentally, over 70% of the adsorption occurs in the first 30 hours. Unfortunately there is no comparison with a model in which only surface sorption is considered that would have provided an indication of the importance of including intra−particle transport nor was there information on the particle sizes used. Given the assumptions of the initial model (spherical porous particles with a presumably homogeneous solid phase interiors), and no consideration of particle composition variability in the model, a logical first step would be the conduct of carefully crafted laboratory experiments to validate the model. **With the lack of such information or any indication of the validation of the sorption model using real particles before application to the proposed modeling effort, it is difficult to assess the potential significance of the work in contributing a significant advance in understanding mercury dynamics in San Francisco Bay or elsewhere for that matter. In this reviewer's opinion the contribution of the intra−particle sub−routine to increase accuracy in modeling the biogeochemical dynamics of mercury in the Bay would be of marginal value.**
Rating	**fair**

Feasibility

Is the approach fully documented and technically feasible? What is the likelihood of success? Is the scale of the project consistent with the objectives and within the grasp of authors?

Comments	**The modeling effort as proposed seems well within the capability of the PIs from a strictly modeling and computational perspective. However there are many gaps in the PIs acknowledgement of the full range of biogeochemical processes that would have to be considered in any such model if the goal is to describe mercury behavior in the environment. For example in the description of Task 2, there is discussion of the need to potentially apply speciation models such as PHREEQC to model the aqueous phase speciation, but, to simplify the model, the model will just consider the "reduction−oxidation reactions among zerovalent and divalent mercury, the reversible methylation reaction between methylated and divalent mercury, and sorption−desorption of both methylated and divalent mercury to porous particle surfaces in both the suspended state and the deposited state." However the [Hg+2] concentration (activity?) is strongly influenced by both inorganic and organic ligands and thus it is not clear how or where data on this species will be arrived at. The statement that "In all reactions the divalent mercury is understood to represent the available divalent mercury because the various complexes that form are not all available to reaction" may not be the case and more importantly does little to indicate what the available divalent mercury is or how it is to be determined for inclusion in the model.** **It is not clear whether the dynamic recycling of biogenic particles, that are important absorbers of mercury, are to be included in the model and, if so, how. Certainly biodegradation and formation of the particles themselves on a variety of timescales in both the water column and especially at the sediment−water interface in shallow ecosystems have been shown to be extremely important in the transport to, and fate within, the sediments for particle−reactive elements like mercury. The model appears to treat all particles as homogeneous absorbers of constant composition over a range in**

particle sizes. How will heterogeneity of composition, biological activity and size distributions in the sediment beds of the Bays be dealt with and at what resolution? Mention is made of the use of a single sediment layer to represent the "active" sediment layer conceptually described in Bale (2000) but unfortunately I am not familiar with this paper and am not clear how they intend to define it. Part of my confusion may also arise from a reference to equation 6 that I believe is actually equation 4. If that layer is similar to the very thin layer (millimeters) of generally unconsolidated sediments typically participating in periodic resuspension/sedimentation events, the nature of the particles and their sorption desorption behavior can be expected to be very different from those in the underlying consolidated sediment bed.

With respect to Task 3, what size classes will be incorporated into the sediment transport sub−model and is the assumption of spherical particles a reasonable one with respect to sediment transport as well as the sorption−desorption processes to be modeled above? The assumption of homogeneous sediments in the horizontal direction is also troubling. How is the sediment transport sub−model to be verified?

In general, given the uncertainties in various phases of the modeling effort, is it reasonable to expect the introduction of an intra−particle modeling sub−routine in the modeling of mercury distributions in the two bays to measurably improve our understanding of mercury transport and methylation in the bays? In this reviewers opinion this seems unlikely. On the other hand the introduction of a more sophisticated intra−particle transport process model is of value in its own right, but needs to first focus on exploration of the validity of the application of the model to natural particles in well−crafted experiments before extrapolation to the field and justifying the benefit of inclusion in already complicated and uncertain

	models describing the dynamics of mercury in coastal ecosystems.
Rating	fair

Monitoring

If applicable, is monitoring appropriately designed (pre−post comparisons; treatment−control comparisons)? Are there plans to interpret monitoring data or otherwise develop information?

Comments	The final model is to be verified using historical field data but how this is to be accomplished is not clear or over what temporal and spatial scales the model will be validated. However the PIs note correctly that the effort in Task 4 is designed as a learning experience to better refine the model and further understand “what to model”. Unfortunately I have not been convinced that inclusion of an intra−particle adsorption kinetic model as proposed is the next best step in this process.
Rating	very good

Products

Are products of value likely from the project? Are contributions to larger data management systems relevant and considered? Are interpretive (or interpretable) outcomes likely from the project?

Comments	Not likely given the limitations and deficiencies in the proposed research.
Rating	fair

Additional Comments

Comments	None

Capabilities

What is the track record of authors in terms of past performance? Is the project team qualified to efficiently and effectively implement the proposed project? Do they have available the infrastructure and other aspects of support necessary to accomplish the project?

Comments	**With respect to the modeling itself the PIs seem to be extremely well qualified. However placing the model effort in a realistic framework such that the stated goals would be achieved is not apparent and necessary to this reviewer.**
Rating	**good**

Budget

Is the budget reasonable and adequate for the work proposed?

Comments	**As far as I can tell. Not sure what level of efforts the stated salaries represent given the rather awkward budget terminology used. Does 1 month of summer salary @ 50% = 0.5 months of summer salary? Does the varying amount of graduate student salary for the different tasks reflect different levels of effort and if so why not state the time required for each task explicitly?**
Rating	**very good**

Overall

Provide a brief explanation of your summary rating.

Comments	**While the PIs are apparently excellent modelers there is a lack of a) a justification for the importance of what they propose, b) an appropriate approach to demonstrate the significance and accuracy of the proposed intra−particle modeling, and c) an appropriate integration with the known biogeochemical processes in the modeling effort that would significantly advance the understanding of mercury**

	dynamics in the bays.
Rating	**fair**

Technical Review #3

proposal title: Processes and particle−associated fate and transport of mercury: quantification tools for risk assessment

Review Form

Goals

Are the goals, objectives and hypotheses clearly stated and internally consistent? Is the idea timely and important?

Comments	**Modeling tools that better capture environmental processes are certainly welcome and the PI's objectives in this field as directed at Hg fate and transport modeling are laudable and clearly stated. Developing novel mathematical frameworks for complex natural processes is always a timely and important idea. However (and this proposal may fall under this caveat) adding complexity where it is not justified or testable and/or adds more fitting parameters and/or is computationally intractable does not take us in the right direction.**
Rating	**good**

Justification

Is the study justified relative to existing knowledge? Is a conceptual model clearly stated in the proposal and does it explain the underlying basis for the proposed work? Is the selection of research, pilot or demonstration project, or a full−scale implementation project justified?

Comments	**Addressing intra−particle diffusion is justified for certain contaminants (e.g. such models better describe the uptake, distribution, and desorption hystesis of hydrophobic PCB's or PAH's into organic materials such as algal cells or lipid layers), however the case for hydrophilic, charged species is much less clear. The justification for adding intra−particle modeling**

	sub−routines to fate models of surface reactive contaminants needs to be improved. **I agree that this type of modeling is valuable in an exploratory sense, running sensitivity scenarios to rank effects and focus future modeling efforts.** **The authors present a good summary of the existing modeling frameworks but comparable detail on Hg geochemistry is lacking.**
Rating	**good**

Approach

Is the approach well designed and appropriate for meeting the objectives of the project? Is the approach feasible? Are results likely to add to the base of knowledge? Is the project likely to generate novel information, methodology, or approaches? Will the information ultimately be useful to decision makers?

Comments	**I suspect the reliance on simple partitioning models for particle surface uptake is doomed to failure – and I think the PIs must recognize this. They express the need for more accurate mechanistic model representations of Hg particle partitioning, yet they ignore a huge body of evidence on solution and surface complexation of Hg. These very real processes cannot be tossed aside. Chemical processes at the particle surface and in solution (ligand exchange; sulfide, thiol, chloride, hydroxide complexation/bonding; precipitation) control the speciation and distribution between "solution" and particle surface. Electrostatic (charge) processes also play a critical role and have been described in various double and triple layer models. pH is a controlling state variable. Divalent, un−complexed Hg exists, but is vanishingly small in natural systems with even trace amounts of DOC and other ligands. The model would need to consider, not just the charged species, but neutral chloride and sulfide complexes that have been shown to be more bioavailable and by inference, better able to move**

across membranes. Sulfide speciation of Hg is very complex and HgS (cinnabar) is but one of many potential reactive solid phases. The proposal does not address these issues. The focus here is clearly on the mathematics and it appears that some of the complexities of Hg biogeochemistry have been lost.

Treating an environmental particle as a porous sphere has significant limitations. Natural particles are often coated with DOM or exist as complex micro−aggregates. Biologically sourced particles/membranes will behave quite differently. Active biological acquisition pathways exist for many metals, though no such pathway has yet been documented for Hg or MeHg. How the model parameterizes particle porosity and tortuosity is vague.

Reduction of Hg++ and evasion as Hgo is an important mechanism in many aquatic systems – it cannot be ignored in models. I have doubts whether methylation of Hg can be modeled as described. In contrast to the authors statement, sulfate is limiting in many environments. Iron becomes a factor through its complexation of sulfide. The PIs do not acknowledge the complex suite of factors affecting sulfate reduction and Hg−methylation and therefore the models promulgated are deficient. This is not to say that existing models perform much better, just that the proposed approach is not capturing the state−of−knowledge.

Hg−binding to sites on particles is a competitive process with both trace and major cations competing for many of the same sites. The PI's state that these various processes will be thoroughly tested, but do not explain how.

Details of how Hg−specific data will be developed to parameterize the models are missing. This information is clearly essential to the success of the study, yet the resources do not seem to be there to provide this

	data. **The sediment transport Task is very poorly developed. There are many holes left unfilled and “modules” ill−described.** **Demonstration−simulations in Task 4 are very poorly developed and described. No clear testable hypotheses are stated.**
Rating	**good**

Feasibility

Is the approach fully documented and technically feasible? What is the likelihood of success? Is the scale of the project consistent with the objectives and within the grasp of authors?

Comments	**I believe the PIs have proposed a nearly intractable computational paradigm. Even with modern computing capabilities the models will need to be substantially simplified to be run. It’s not clear that after the required restructuring that the stated goals will be achievable.** **The complexity of the model suggests that validation will be difficult. Application to the California bay sites is substantially jumping the gun and the PIs must step back several steps to attempt validation in a much more controlled, and simplified environment. Whether this is in lab scale reactors, or flumes to bring in more particle transport, these tests are critical to address relevance and efficacy. At some point as the modeling reaches maturity and laboratory validation performed, a real field experiment (e.g. a tracer injection) should be considered.**

	Though in theory a size−segregated modeling approach seems reasonable to pursue, in the real world particle−size becomes a fuzzy concept. Aggregation, density and shape differences, particle chemistry all conspire to complicate not only particle transport, but also contaminant−particle interaction. Only a few of the hundreds of studies examining the relationship between particle−size and contaminant association have shown surface area effects in natural systems. Existing data on initiation of motion criteria for sediments and models of bed−sediment and surface sediment transport are at best primitive and most wholly inadequate of fate and transport modeling of contaminants.
Rating	**good**

Monitoring

If applicable, is monitoring appropriately designed (pre−post comparisons; treatment−control comparisons)? Are there plans to interpret monitoring data or otherwise develop information?

Comments	**This proposal does not include a traditional monitoring component.**
Rating	**not applicable**

Products

Are products of value likely from the project? Are contributions to larger data management systems relevant and considered? Are interpretive (or interpretable) outcomes likely from the project?

Comments	**It’s likely that the modeling framework and code that evolves from this study will add value to contaminant fate assessment. Unfortunately validation of the models will be difficult – so efficacy will be**

	uncertain at best. It's not clear to me that Hg is the best contaminant to apply this approach to – a hydrophobic organic might be better suited. However there should be value irregardless in pushing ahead with increasing the sophistication of the models.
Rating	good

Additional Comments

Comments	no additional comments.

Capabilities

What is the track record of authors in terms of past performance? Is the project team qualified to efficiently and effectively implement the proposed project? Do they have available the infrastructure and other aspects of support necessary to accomplish the project?

Comments	The PI's have not published nor do they have extensive experience in the area of Hg biogeochemistry. Though not an overriding issue from the modeling side, the unique characteristics of Hg present challenges to those with less trace element background. The PI's do have an impressive record of accomplishment/capability in fluid mechanical modeling and mathematical simulation of complex systems. Given a conceptual framework or geochemical pathway, I have little doubt that the team could develop an advanced mathematical model.
Rating	very good

Budget

Is the budget reasonable and adequate for the work proposed?

Comments	The budget seems wholly inadequate for the effort proposed. The PIs significantly underestimate the time and resources that would be required to effectively carry−out this work.

Rating	**fair**

Overall

Provide a brief explanation of your summary rating.

Comments	**The PI's are attempting to tackle way too much. Anyone of the many Tasks outlined could conceivably require a full project of its own. In attempting to develop more mechanistically accurate environmental models, the PIs have painted themselves into a computationally intractable corner and in part because of the added complexity, have made validation of the models problematic. Validation is critical and not adequately addressed. It's also uncertain whether intra-particle diffusion is relevant for Hg and MeHg and testing of this hypothesis is difficult under the framework outlined in the proposal. Certain tasks are poorly developed which suggest that the PIs did not recognize the complexities of the activity. Lab studies on contaminant movement within particles are needed to validate model. Data on changes in reactivity of particle associated Hg with time could help address the transport issue.**
Rating	**good**